Remarks

Claims 8 - 18 are pending. Favorable reconsideration is respectfully requested.

Applicants wish to extend their appreciation to Examiner Ojurongbe for his thorough examination of this application.

The claimed subject matter is directed to a primer coat for silicone release-coated substrates. As is well known, the primer coats must possess several important characteristics. First, the primer should fill up the pores of the paper substrate so that the subsequently applied silicone will not penetrate into the paper. Second, the primer must adhesively bond to the silicone so that when subsequently attached pressure sensitive adhesive-coated labels and the like are applied, these can be easily removed without also removing the release coating. The adhesion of the silicone to the primer may be assessed by conventional rub-off tests, as described in the specification on page 10, and the "migration" test, where the amount of silicone adhering to an adhesive tape is measured.

In the specification, various prior art primers and their disadvantages are discussed. For example, Uwe Skurnia et al. DE 3727078 ("*Skurnia*") is discussed on page 1 at lines 16 - 18, and Maruyama U.S. 4,617,239 ("*Maruyama*") is discussed as JP-A 58/214596. The disadvantages of both these systems is discussed beginning at lines 33 - 38. Thus, Applicants teach not to use foundation coatings containing metal complexes (such as chrome stearates) and unmodified silane-containing polyvinylalcohols.

In view of the above, claim 1 has been amended to recite that Applicants' primer coatings are free of metal complexes. New claim 18 has been added to recite that the primer consist essentially of the 1-alkylvinyl ester-containing silane-modified polyvinyl alcohol and further additives selected from those additives therein listed (from claim 17), which precludes the use of metal complexes as additives, since the Markush group of additives is prefaced by

"selected from the group consisting of". Thus, additives other than those listed are not within the scope of the claim. No issue of new matter is raised by virtue of these amendments.

The claims have been rejected under 35 U.S.C. § 103(a) over *Skurnia* in view of *Maruyama* further in view of Tschirner et al. ("*Tschirner*"). Applicants respectfully traverse this rejection. The *Skurnia* reference is very confusing and difficult to understand. If the rejection over *Skurnia* is maintained, a proper English translation, per the Commissioner's Notice in the Official Gazette, is requested.

As best understood, *Skurnia* is directed to the preparation of release substrates, onto which adhesive labels or adhesive tape may be applied. *Skurnia* employs commercially available silicones for the release coating. The improvement *Skurnia* discloses and claims is providing a "Grundierung" (foundation coat) which itself has a high releasability ("Trennwirkung") (i.e. the foundation coat itself is a release coating), onto which are screen printed ("Siebdruck") a "raster-like" ("rasterartiges") coating of the commercial silicone release agent. In the examples, dots having a mesh number ("mesh-Zahlen") of 80 - 125 are deposited.

As the low release value foundation coat, *Skurnia* discloses an isocyanate crosslinked polyvinylacetate ("isocyanatvernezte Polyvinylacetate") and a chrome stearate complex with a small (unspecified) amount of polyvinylalcohol as a film former. Such chrome stearate release coatings have been used for decades.

As the specification makes clear, a major problem with prior art "pre-coatings" (coatings applied to the paper prior to applying the silicone release coat) is that they do not adhere sufficiently to the silicone release coat. As a result, the silicone may "migrate" ("tear-away") onto the adhesive article applied onto the release coat. This is the case with the products of *Skurnia*. The "pre-coat" or "foundation coat" as *Skurnia* describes it, is itself a coating with low surface energy - an efficient release coating itself. Because of this, the silicone coating applied thereon cannot be bonded effectively, and significant migration will occur. If migration thus occurs to any significant degree, and the silicone release coating were a continuous one, the

entire adhesive surface of the label or the like could come away with a silicone coating over all or a substantial part of the adhesive surface. The label would then not stick firmly to the article on which it is to be fastened. Note the migration test at the top of page 10 of the present specification describing this phenomenon. *Skurnia* partially solved this problem by use of two release layers: a first, continuous layer of modest releasability (chrome stearate complex with PVA film former), onto which a raster of a much better release agent (silicone) is applied. Because the silicone is applied in the form of isolated "dots", the releasability will be a function of both the first and second coats, since the first coat is exposed in areas between the silicone "dots." The releasability will thus be compromised: less than if a continuous silicone coating is used. The migration problem is not prevented by this technique. Rather, significant migration is to be expected due to the releasability of the first coat. However, when the silicone does migrate to the adhesive of the label, it must do so in the form of dots, leaving extensive areas of the adhesive "undotted." For example, if the dots of the silicone release add up to 40% of the release coating surface area and there is 100% migration, the adhesive surface of the label will still have 60% of adhesive surface.

Since the first coat of *Skurnia* must have a low release value, but is otherwise of unspecified properties, the least expensive compositions would be used by one skilled in the art for this purpose. The chrome stearate complex release coatings have been used for a long, long time, are available from numerous sources, and at low cost. One would not be motivated to replace these economical release coats with a more expensive product, such as a silane-modified PVA. Furthermore, there is no indication anywhere that a silane-modified PVA would provide the low release force necessary for the raster printed two layer release coat of *Skurnia*. Note that in *Skurnia,* the foundation coat must itself be a release coat.

Maruyama discloses silane-modified PVA for use as a paper coating. The *Maruyama* patent is assigned to Kuraray, a Japanese company which supplies numerous types of polyvinylalcohols to the market, under the tradenames MOWIOL® and POVAL®. The silane-modified PVA and by *Maruyama* in Example 1 is the same or substantially the same as Kuraray POVAL-R 1130. This PVA was used by Applicants in Comparative Example 2.

The problems of *Maruyama's* silane-modified PVA are that first, its solution viscosity is high (as shown by the Kuraray article attached) and second, aqueous solutions rapidly increase in viscosity due to condensation of the silanol groups in the polymer. Thus, the polymer is supplied neat, and must be dissolved on site and used immediately. To solve the high viscosity problem, Kuraray has recently introduced a new silane-modified PVA, POVAL-R 3109. However, the lower viscosity silane-modified PVA is not believed to have been available at the time Applicants' invention was made. *Maruyama* indicates that his silane modified PVA rapidly gels and reacts with the hydroxyl groups of the cellulosic content of the paper, providing strong adhesion and pore-filling capacity. However, *Maruyama* is silent about the releasability of this coating. Since the coatings are said to have improved printing characteristics, it would be assumed by one skilled in the art that releasability is very low; otherwise the print would be easily rubbed off or smeared.

For the reasons given above, one skilled in the art would not be motivated to substitute a *Maruyama* silane-modified PVA for the chrome stearate complex foundation release coat of *Skurnia*, because 1) its high solution viscosity; 2) its higher cost; and 3) lack of any indication that the coating would have a low release value. For these reasons alone, the rejection of the claims should be withdrawn.

Maruyama is directed to silane-modified polyvinyl alcohols suitable for coating paper stock for use in preparing offset printing plates and to increase paper strength. The copolymers of *Maruyama* are prepared by copolymerizing polyvinylacetate and an unsaturated silane monomer followed by hydrolysis to the polyvinyl alcohol (PVA1). However, it is well known in the art that the silane moieties in *Maruyama's* polyvinyl alcohols, are subject to hydrolytic condensation reactions:

$$\text{polymer}—Si(OCH_3)_3 \xrightarrow{H_2O}$$

$$\text{polymer}—Si(OCH_3)_2—O—Si(OCH_3)_2—\text{polymer} + 2CH_3OH$$

This initial chain-extending reaction may be followed, due to the presence of still-unreacted alkoxy groups, by branching and crosslinking. Furthermore, the silyl methoxy groups are reactive with the hydroxyl groups of PVA1. All these increase the viscosity of the polymers greatly, and thus incorporation of such silane groups into polyvinyl alcohols is known to decrease storage stability, manifested by a marked increase in viscosity. Moreover, this is not a physical phenomenon which is reversible, but a chemical phenomenon forming more favorable chemical bonds, and essentially irreversible. *Maruyama* was first published in 1983 (Japan), and issued as the U.S. Patent in 1986. The Japanese publication was *ca.* 20 years before the present invention.

Tschirner is not directed to polyvinylalcohols, but rather is directed to specially modified polyvinyl acetals. These polyvinyl acetals are prepared by acetalizing (for example with butyraldehyde) a polyvinyl alcohol copolymer derived from vinylacetate and 1-alkylvinyl acetate. Following hydrolysis of the latter, reaction with the aldehyde takes place under acid catalysis. The polyvinyl acetals are useful as binders for printing inks.

One problem with the preparation of low molecular weight polyvinyl acetals is that the corresponding low molecular weight polyvinyl alcohol starting materials tend to gel in aqueous solution, in a manner similar to gelatin. This is not a result of chemical reaction, but rather one of association and hydrogen bonding of polar groups (polyvinyl alcohols have one polar, hydrogen bondable OH group per vinyl group, thus, many of these groups). This association is reversible, and just as does gelatin, such products liquify at higher temperatures, *e.g.*. 50°C. *See Tschirner* at column 1, lines 52 - 58. This room temperature gellation of PVA1 was improved by incorporating a 1-alkylvinyl acetate moiety into the PVA1, followed by acetalization. As is the case with gelatin, gellation is only a problem at high concentration.

However, *Tschirner* is not directed to use of PVA1 as a primer, nor for any purpose whatsoever except as a precursor to polyvinyl acetals. *Tschirner* surprisingly discovered that the viscosity of polyvinyl acetals formed from the 1-alkyl vinyl acetate-modified PVA1's was independent of the vinyl alcohol content over a wide range, and that the resulting polyvinyl

acetals, at the same molecular weight as acetals formed from homopolymeric PVA1, had a lower solution viscosity.

Thus, the only teaching one skilled in the art would glean from *Tschirner* is that in preparing polyvinyl acetals where low viscosity is desired, one should start with 1-alkylvinyl acetate-modified PVA1s rather than PVA1 homopolymers. *Tschirner* was first published in 1998, some 5 years prior to Applicants' invention.

The problems addressed by the subject invention and those addressed by *Maruyama* and *Tschirner* are completely different. *Maruyama* wished to minimize penetration of PVA1 into paper by supplying a reactive PVA1 which reacts not only with cellulosic hydroxyl groups, but also with hydroxyl groups of PVA1 itself. *See Maruyama* at column 9, lines 8- 25. Thus, the polymer gels rapidly at the paper interface (containing huge numbers of hydroxyl groups), preventing its migration into the paper. This reaction of hydrolyzable silyl groups with PVA1 hydroxy groups is also responsible for the aqueous instability noted earlier, which all skilled in the art are aware of, and to which reaction *Maruyama* himself attests.

Tschirner, on the other hand, is only marginally interested in PVA1s at all - only as a starting material for polyvinyl acetals. For this purpose, he notes that avoiding room temperature gellation is desirable (but certainly not necessary, as he very easily made polyvinyl acetals from conventional PVA1s as well (see all his comparative examples)). However, if *Tschirner's* discussion of avoiding polyvinyl alcohol gellation is the only isolated teaching derived from him, it is clear that <u>any modification of PVA1 copolymers which would be expected to generate viscosity increases would be directly against his teaching</u>.

However, *Maruyama* indicates that his reactive silane groups react with the PVA1 OH groups (it has been known for many years that such silane groups react with each other - the basis for moisture curable silicone caulks and sealants), and thus one skilled in the art would not be motivated to add a silyl group of *Maruyama* to a PVA1 of *Tschirner*, since the expected result would be an unstable polymer which exhibits considerable viscosity increase in aqueous solution,

and would gel at even lesser concentrations than *Tschirner's* PVA1 or conventional homopolymeric PVA1.

Thus, not only would one skilled in the art not be motivated to add hydrolyzable silane groups to *Tschirner's* PVA1, but moreover, the result is not the expected result, contrary to the holding in *KSR*, for example. Here, as shown by the examples and comparative examples, the silane-modified inventive PVA1's of the subject invention showed virtually no viscosity increase upon storage. The viscosity only increased from 28.1 mPa·s to 28.3 mPa·s over two weeks. This is an increase of only 0.7%, and being so small, is even hard to measure and differentiate accurately! Yet the very similar *Maruyama* silane-modified homo-PVA1 showed a viscosity increase of 260%, an increase of a factor of 370 (260 ÷ 0.7)! One skilled in the art would expect that a silane-modified 1-alkylvinyl/vinyl alcohol copolymer would show a similar increase in viscosity, due to the fact that both contain the same reactive silane group, and a huge number of PVA1 hydroxyl groups as well. However, surprisingly that was not the case.

Moreover, the acetalization of PVA1 to polyvinyl acetals, for reasons of economy (smaller storage tanks, reactors, pumps, etc.) employs concentrated PVA1 solutions. Note in *Tschirner's* examples, that PVA1 was supplied as a 20% solution. Primer coatings do not employ such concentrated solutions. For example, the viscosity measurements reproduced in the tables used a concentration of PVA1 which is only about half (11%) than that employed by *Tschirner*. At this lower PVA1 content, gellation is not a problem. For example, if the directions on a package of gelatin (e.g. JELLO™) are not followed, and twice as much water is added, no gel will be formed - only a liquid.

It is also noted that *Maruyama* (*See* column 9) discloses that it is necessary that his product gel at the surface of the substrate to prevent penetration. Contrary to this, the teaching relied upon from *Tschirner* is to provide PVA1s which do not gel. These objectives are diametrically opposed, and therefore uncombinable. One would not modify a *Maruyama* silane-modified PVA1 in a manner so as to prevent gelling which *Maruyama* desires.

However, in addition, the present inventor's have achieved surprising and unexpected results. After seven days of storage, the silicone release coat applied over the primer coat showed a much higher scratch resistance than either the silane-modified PVA1 of *Maruyama* or a conventional PVA1. This result is neither taught nor suggested by *Maruyama*, *Tschirner*, or their combination.[1]

The difference in scratch resistance is striking, and cannot be due to viscosity effects alone, since at seven days, the ratio of silane-modified PVA1 viscosity to subject invention PVA1 viscosity is only about 2:1 (61.7 mPa·s v. 28.2 mPa·s). Both these are relatively low viscosity, about that of the vegetable oils (raw linseed oil, *ca.* 28 mPa·s; corn oil, *ca.* 72 mPa·s). If anything, the more viscous silane-modified PVA1 of *Maruyama* would be expected to exhibit <u>less</u> penetration into the substrate (which was *Maruyama's* objective) and thus be <u>more</u> efficient as a primer. Instead, the results show that Applicants' <u>less viscous</u> primer is superior. Why is this? Why should it be so? This result is certainly surprising and unexpected. Withdrawal of the rejection of the claims on this basis is further solicited.

Providing a primer which allows both good adhesion to silicone release coatings as well as allowing rapid cure of the release coatings is a long-felt need, as attested to be the first few pages of the specification. The invention of *Maruyama* had been known to the public for some 20 years prior to Applicants' invention. *Tschirner* was known to the public some five years prior to the present invention. Despite this knowledge, prior to Applicants, no one had attempted to produce a primer of Applicants based on these teachings. The existence of a long felt need and a solution which occurred only many years after the cited references were published is an important secondary consideration which must be considered in assessing patentability. *See, e.g. In re Sernaker*, 217 USPQ 1 (Fed. Cir. 1983).

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[1]The increased scratch resistance is not recited in the claims, but it need not be, when the feature responsible therefore (the use of Applicants' primer) is recited. *See, e.g. In re Merchant*, 197 USPQ 785 (CCPA 1978).

For the foregoing reasons, it is respectfully submitted that one skilled in the art would not be motivated to combine *Maruyama* and *Tschirner*; that the references themselves have opposing goals with respect to gellation; that unexpected and surprising results have been achieved, not an expected result; and that Applicants' invention has resolved a long felt need many years after the references' publications. Withdrawal of the rejection for all these reasons is respectfully solicited.

To reemphasize, *Skurnia* teaches that his foundation coat is itself a coating with high releasability. This is necessary because the even higher releasability silicone is not applied continuously over the surface, but as a raster of dots. If the first coat did not also offer at least modest releasability, the PSA article attached to the paper would not be removed, or could be removed only with great difficulty. There is no evidence that the *Maruyama* polymer has any release nature. Thus, one skilled in the art would not be motivated to use it in a *Skurnia* release paper, particularly as it is also more expensive and has application and storage problems.

The effect of addition of 1-alkylvinyl ester moieties to silane-modified PVA is completely unpredictable. Silane-modified PVA is unstable in aqueous solution, and has a high initial solution viscosity as well. *Tschirner* teaches that 1-alkylvinyl-modified PVAs have stable solution viscosities. However, there is no teaching that silane-modified, 1-alkylvinyl-modified PVAs would also have a stable solution viscosity. The reason for this is that silane-modified PVAs of <u>all</u> types still contain hydrolytically unstable and reactive silyl groups. Why would the presence of a 1-alkylvinyl group prevent these groups from reacting?

Silane-modified PVAs are known to have viscosity problems due to the reactivity of the silyl groups. One skilled in the art would certainly be motivated to improve the storage stability. How would one do this, however? One skilled in the art might be motivated to replace the reactive silyl groups or a portion thereof with less reactive, perhaps inert silyl groups such as a trimethylsilyl group. However, this would interfere with reaction with the paper. One might choose to use a lesser quantity of reactive groups. This again would interfere with its performance. Would one be motivated to add 1-alkylvinyl units to such a polymer? I think not.

There is no reason why one skilled in the art would expect any change in reactivity of alkoxysilyl or silanol groups by addition of a further monomer which is essentially inert. Why not copolymerize ethylene or propylene, or styrene, methylacrylate or methylmethacrylate? One skilled in the art is simply not directed by *Tschirner* to include 1-alkylvinyl groups in a silane-modified PVA. There is no reason to do so, and no expected result.

For all the above reasons, withdrawal of the rejection of the claims over *Skurnia* in view of *Maruyama* further in view of *Tschirner* is respectfully solicited.

It is noted that claim 16 has been amended to recite that the silicone release coating is a continuous film, as shown in all the examples. *Skurnia* teaches away from the use of a continuous film, and requires a raster of discontinuous dots or islands of silicone. Claim 16 is separately patentable.

Applicants submit that the claims are now in condition for Allowance, and respectfully request a Notice to that effect. If the Examiner believes that further discussion will advance the prosecution of the Application, the Examiner is highly encouraged to telephone Applicants' attorney at the number given below.

Please charge any fees or credit any overpayments as a result of the filing of this paper to our Deposit Account No. 02-3978.

Respectfully submitted,

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Attachment: Page 1 on Kuraray Poval-R from Kuraray web site.

Special Grades

KURARAY R-POLYMER

1. Introduction

KURARAY R-polymer is water-soluble polymer, which has peculiar functional groups, i.e., silanol groups, in their molecular structures. The silanol groups are reactive with inorganic substances such as silica or alumina. R-polymer can be easily cross-linked by changing PH of their aqueous solutions or by mixing with inorganic substances and can form water resistant films.
R-polymer is mainly used as a binder for inorganic substances and as surface coating agent for organic materials containing inorganic substances such as paper.

2. Grades and specifications

GRADES AND SPECIFICATIONS

Grades	R-1130	R-2105	R-3109
Appearance		pale yellow granular powder	
Solubility	water soluble	water soluble	water soluble
Volatiles(% max)	5.0	5.0	5.0
Viscosity(cps)*1	20-30	4.5-6	9-12
Remarks	*1) Viscosity in cps of 4% aqueous solution at 20℃ is determined by Brookfield Viscometer.		

The conventional R-polymer, such as R-1130, has some difficulties in its usage for paper coating since they have comparatively higher viscosity. KURARAY R-polymer R-3109 is newly developed improving such difficulties by adjusting their viscosity and it is easy to use them in the actual operation of the paper coating.

3. Characteristics of R-polymer

Reactivity with inorganic substances
R-polymer has high reactivity with a variety of inorganic substances and forms water